Possible vacancy induced supersolid in ^4He

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In the Ginsburg Landau (GL) theory developed by the author to study possible supersolid in ^4He, there are two parameters: v is the repulsive interaction between the normal component and the local superfluid mode, g is a periodic changing chemical potential for the local superfluid mode. If $g = 0$, the normal solid has a particle-hole symmetry. If g is negative (positive), the normal solid is a vacancy (interstitial) type normal solid. Taking the P-H symmetric normal solid as a reference state, we show that when g is sufficiently negative, the vacancy induced supersolid is the ground state. For comparison, we also discuss interstitial induced supersolid. The microscopic origins of v and g are discussed. Experimental implications are given.

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1. Introduction A solid can not flow. It breaks a continuous translational symmetry into a discrete lattice translational symmetry. While a superfluid can flow even through narrowest channels without any resistance. It breaks a global $U(1)$ phase rotational symmetry. A supersolid is a state which breaks both kind of symmetries. The possibility of a supersolid phase in ^4He was theoretically speculated in 1970 [1]. Over the last 35 years, a number of experiments have been designed to search for the supersolid state without success. However, recently, by using torsional oscillator measurement, a PSU group lead by Chan observed a marked $1 \sim 2\%$ superfluid component of solid ^4He at $\sim 0.2K$ above $25bar$, both when embedded in Vycor glass and in bulk ^4He [2]. Very recent specific heat measurements found a excessive specific heat peak around the putative supersolid onset critical temperature $\sim 100\ mK$ [3]. The authors suggested that the supersolid state of ^4He maybe responsible for the superfluid component. The PSU experiments rekindled extensive both theoretical [4, 5, 6, 7] and experimental [8, 9] interests in the still controversial supersolid phase of ^4He. There are two kinds of complementary theoretical approaches. The first is the microscopic numerical simulation [4]. The second is the phenomenological approach [6, 7]. At this moment, despite all the theoretical work, there is no consensus at all on the interpretation of PSU's experiments. In [7], the author constructed a phenomenological Ginsburg Landau (GL) theory to study all the possible phases and phase transitions in ^4He and map out its global phase diagram from a unified framework. In this paper, by using the GL theory developed in [7], we study the possible microscopic mechanism leading to the supersolid and analyze carefully the conditions for the existence of the supersolid. We find that SS-v is more likely than SS-i. However, in order to be complete, we study both cases on the same footing. It is also constructive to compare SS-v with SS-i even though the SS-i is unlikely to be relevant to the Helium 4 system. We call vacancies induced supersolid as SS-v, interstitials induced supersolid as SS-i. When the two kinds of SS show different properties, we treat them differently, when they share the same properties, we treat them just in the same notation SS.

2. Ginzburg-Landau theory of a supersolid: Let's start by briefly reviewing the GL theory of a supersolid in [7]. The density of a normal solid (NS) is defined as $n(\vec{x}) = n_0 + \sum'_{\vec{G}} n_{\vec{G}} e^{i\vec{G}\cdot\vec{x}} = n_0 + \delta n(\vec{x})$ where $n^*_{\vec{G}} = n_{-\vec{G}}$ and $\vec{G}$ is any non-zero reciprocal lattice vector. In a normal liquid (NL), if the static liquid structure factor $S(k)$ has its first maximum peak at $\vec{k}_n$, then near k_n, $S(k) \sim \frac{1}{r_n + c(k^2-k_n^2)^2}$. If the liquid-solid transition is weakly first order, it is known that the classical free energy to describe the NL-NS transition is [10]:

$$\begin{aligned} f_n &= \sum_{\vec{G}} \frac{1}{2} r_{\vec{G}} |n_{\vec{G}}|^2 - w \sum_{\vec{G}_1,\vec{G}_2,\vec{G}_3} n_{\vec{G}_1} n_{\vec{G}_2} n_{\vec{G}_3} \delta_{\vec{G}_1+\vec{G}_2+\vec{G}_3,0} \\ &+ u_n \sum_{\vec{G}_1,\vec{G}_2,\vec{G}_3,\vec{G}_4} n_{\vec{G}_1} n_{\vec{G}_2} n_{\vec{G}_3} n_{\vec{G}_4} \delta_{\vec{G}_1+\vec{G}_2+\vec{G}_3+\vec{G}_4,0} + \cdots \end{aligned} \quad (1)$$

where $r_{\vec{G}} = r_n + c(G^2 - k_0^2)^2$ is the tuning parameter controlled by the pressure or temperature. Note that because the instability happens at finite wavevector, Eqn.1 is an expansion in terms of small parameter $n_{\vec{G}}$ alone, it is *not a gradient expansion* ! Note that the average density n_0 does not enter in the free energy. The GL parameters w and u_n may be determined by fitting the theoretical predictions with experimental data.

Of course, the Superfluid (SF) to Normal Liquid (NL) transition at finite temperature is the 3d XY transition described by:

$$f_\psi = K|\nabla\psi|^2 + t|\psi|^2 + u|\psi|^4 + \cdots \quad (2)$$

where ψ is the complex order parameter and t is the tuning parameter controlled by the temperature or pressure. The GL parameters K, t, u may be determined by fitting the theoretical predictions with experimental data.

The coupling between $n(\vec{x})$ and $\psi(\vec{x})$ consistent with all the symmetry can be written down as:

$$f_{int} = g\delta n(\vec{x})|\psi(\vec{x})|^2 + v(\delta n(\vec{x}))^2|\psi(\vec{x})|^2 + \cdots \quad (3)$$

where $\delta n(\vec{x}) = n(\vec{x}) - n_0 = \sum'_{\vec{G}} n_{\vec{G}} e^{i\vec{G}\cdot\vec{x}}$. Note that the average density n_0 does not enter in the interaction.

Eqn.3 is an expansion in terms of two small parameters $\delta n(\vec{x})$ and $\psi(\vec{x})$. The $\cdots$ include higher odd and even powers of $\delta n(\vec{x})$ which are sub-leading to the g and v term. We keep both g and v term in the Eqn.3, because the g term changes the sign, while the v term is invariant under under the Particle-Hole (PH) transformation $\delta n(\vec{x}) \rightarrow -\delta n(\vec{x})$, so the sign of g makes a difference ! Due to the two competing orders between the solid and the superfluid, we expect v to be always positive and is an increasing function of the pressure p. On the other hand, we can view g as a *periodic* chemical potential *with average zero* acting on ψ. it is easy to see the coupling is attractive $g_v < 0$ for vacancies, but repulsive $g_i > 0$ for interstitials. If $g = 0$, the C-NS has the P-H symmetry, let's call this kind of PH symmetric C-NS as NS-PH (Fig.1). In general, $g \neq 0$, so there is no particle-hole symmetry in the C-NS, there are also two kinds of C-NS: (1) vacancy like C-NS (named NS-v) where the excitation energy of a vacancy is lower than that of an interstitial. (2) interstitial like C-NS (named NS-i) where the excitation energy of an interstitial is lower than that of a vacancy. We expect that in contrast to v, g is an intrinsic parameter of solid Helium 4 which depends on the mass of a ^4He atom and the potential between the ^4He atoms, but not sensitive to the pressure p.

The GL equations 1,2,3 are invariant under both the translational symmetry $\vec{x} \rightarrow \vec{x} + \vec{a}, n(\vec{x}) \rightarrow n(\vec{x} + \vec{a}), n(\vec{G}) \rightarrow n(\vec{G})e^{i\vec{G}\cdot\vec{a}}, \psi(\vec{x}) \rightarrow \psi(\vec{x} + \vec{a})$ and the global $U(1)$ symmetry $\psi \rightarrow \psi e^{i\theta}$. In a NL, $n_{\vec{G}} = 0, <\psi>= 0$. In a SF, $n_{\vec{G}} = 0, <\psi>\neq 0$. In a NS, $n_{\vec{G}} \neq 0, <\psi>= 0$, while in a supersolid (SS), $n_{\vec{G}} \neq 0, <\psi>\neq 0$. From the NL side, one can approach both the NS and the SF. Inside the NL, $t > 0$, ψ has a gap, so can be integrated out from Eqn.3, we recover the NS-NL transition tuned by $r_{\vec{G}}$ in Eqn.1. Inside the NL $< n(\vec{x}) >= n_0$, the density fluctuations of $\delta n(x)$ is massive, so can be integrated out from Eqn.3, then we recover the NL to SF transition tuned by t in Eqn.2.

3. The effects of the coupling constants g and v : Inside the NS, the translational symmetry is already broken, we can simply set $\delta n(\vec{x}) = n(\vec{x}) - n_0 = \sum'_{\vec{G}} n_{\vec{G}} e^{i\vec{G}\cdot\vec{x}}$ and put it into Eqn.3 to look at the effects of g and v. Imagine that at a given pressure $p > p_{c1} \sim 25\ bar$, if tuning $g \rightarrow 0$, but keeping v intact, then the normal solid becomes an asymptotically P-H symmetric normal solid (NS-PH) in Fig.1. It is easy to see that v term increases the mass t of ψ at $\vec{k} = 0$ in the NL to $t_{NS-PH} = t + v\sum'_{\vec{G}} |n(\vec{G})|^2$ in the NS-PH. Taking the temperature scale $t = T - T_{XY}$ as the reference scale, then $t_{NS-PH} = T + \Delta(p)$ where $\Delta(p) = v\sum'_{\vec{G}} |n(\vec{G})|^2 - T_{XY} > 0$ is the $T = 0$ gap for the local superfluid mode ψ at $\vec{k} = 0$ in the NS-PH. Because as the pressure p increases, the repulsive interaction v also increases, so it is reasonable to assume that $\Delta(p)$ is a monotonic increasing function of p (Fig.1a). Because

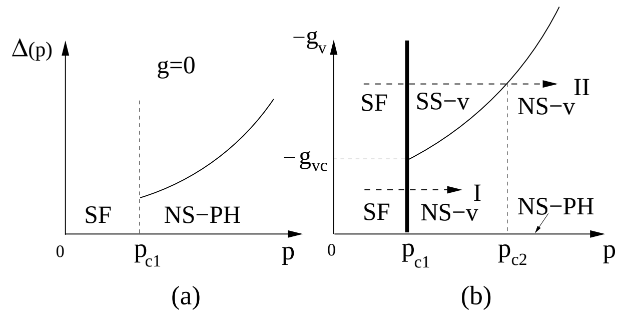

FIG. 1: (a) The gap $\Delta(p)$ of the local fluctuating superfluid mode ψ in the P-H symmetric commensurate normal solid (NS-PH) which exists only at $g = 0$. We assume it is a monotonic increasing function of the pressure p. (b) The zero temperature phase diagram of g_v verse the pressure p. The NS-PH only exists at $g_v = 0$. Any $g_v \neq 0$ will transfer the NS-PH into the NS-v. If the experimental is along the path I, then it is a direct 1st order SF to NS-v transition, if it is along the path II, it is a 1st order SF to SS-v, then a 2nd order SS-v to NS-v transition.

it is a first order transition across p_{c1}, just like the roton gap $\Delta_r > 0$ remains finite just before the first order transition, $\Delta(p)$ also remains finite just after the first order transition, namely, $\Delta(p^+_{c1}) > 0$ (Fig.1a).

Taking this NS-PH as a reference state, we then gradually turn on g and see how the ground state evolves. In the presence of the periodic potential of $n(x)$ lattice, ψ forms a Bloch wave, the u interaction of ψ in Eqn.2 certainly favors extended Bloch wave over strongly localized Wannier state. In principle, a full energy band calculation incorporating the interaction u is necessary to get the energy bands of ψ. Fortunately, qualitatively important physical picture of GL Eqns.1, 1, 3 can be achieved without such a detailed energy band calculation. Neglecting the self interaction u which is not important in the extended Bloch states and taking $g \neq 0$ as a small parameter, a perturbative estimate on the eigenenergy $\epsilon_\mu(0)$ at the origin $\vec{k} = 0$ up to the third order g^3_μ is:

$$\epsilon_\mu(0) = t - g^2_\mu P\frac{n^2(\vec{G})}{K_\mu G^2} + g^3_\mu \sum'_{\vec{G}_1}\sum'_{\vec{G}_2} \frac{n(\vec{G}_1)n(\vec{G}_2)n(-\vec{G}_1-\vec{G}_2)}{K_\mu G^2_1 K_\mu G^2_2} + \cdots \quad (4)$$

where the term linear in g vanishes, $\mu = v, i$ stands for vacancies and interstitials respectively and $G = 2\pi/a$ with $a \sim 3.17\mathring{A}$ the lattice constant of the solid ^4He, the $n(\vec{G}) = e^{-G^2a^2/4\alpha}$ can be taken as a Gaussian where α is the width of the Gaussian..

In reality, g_μ maybe large, so we may need to go beyond leading orders. For vacancies, $g_v < 0$, without writing out the coefficients explicitly, the expansion is:

$$\epsilon_v(0) = t - g^2_v - |g_v|^3 - |g_v|^4 - \cdots \quad (5)$$

so all the coefficients have *the same* sign. Assuming the series converges, we can write $\epsilon_v(0) = t - f_v(g_v)$ where $f_v(g_v) \geq f_v(0) = 0$ is a monotonic increasing function of g_v and likely has no upper bound.

For interstitials, $g_i > 0$, without writing out the coefficients explicitly, the expansion is:

$$\epsilon_i(0) = t - g_i^2 + g_i^3 - g_i^4 + \cdots \tag{6}$$

so the coefficients have *oscillating* sign. Assuming the series converges, we can write $\epsilon_i(0) = t - f_i(g_i)$ which holds for any g_i. Because of the oscillating nature of the expansion coefficients, it is hard to judge the nature of the function of $f_i(g_i)$ except that $f_i(0) = 0$.

The different expansion series of $f_v(g_v)$ and $f_i(g_i)$ indicate that quantum fluctuations may favor vacancies over interstitials. However, *for simplicity*, we assume $f_i(g_i)$ is also a monotonic increasing function of g_i, so we can discuss vacancies and interstitials induced supersolids on the same footing.

4. Vacancies induced supersolid (SS-v): The mass of ψ_v was *decreased* to $t_{\psi_v} = T + \Delta(p) - f_v(g_v) = T - T_{SS-v}$ where $T_{SS-v}(p) = f_v(g_v) - \Delta(p)$ (Fig.2). Because $f_v(g_v)$ is a monotonic increasing function of $|g_v|$ and $f_v(0) = 0$, defining a critical value $f_v(g_{vc}) = \Delta(p_{c1})$, then when $|g_v| < |g_{vc}|$, $f_v(g_v) < \Delta(p_{c1})$, the ψ_v mode remains massive, namely $< \psi_v >= 0$. The NS-v remains to be the ground state even at $T = 0$(Fig.1b).

If $|g_v| > |g_{vc}|$, then $T_{SS-v}(p_{c1}) = f_v(g_v) - \Delta(p_{c1})$ is raised above the zero temperature, the SS-v state exists in the Fig.1b. $T_{SS-v}(p_{c1})$ is also proportional to the superfluid density measured in the experiments. The resulting solid is an in-commensurate solid with vacancies even at $T = 0$ whose condensation leads to $< \psi_v > \neq 0$. The SS-v state has a lower energy than the NS-v state at sufficiently low temperature. As the pressure increases to p_{c2}, $T_{SS-v}(p_{c2}) = f_v(g_v) - \Delta(p_{c2}) = 0$ (Fig.1b). Then $f_v(g_v) = \Delta(p_{c2})$, so $T_{SS-v}(p) = \Delta(p_{c2}) - \Delta(p)$ which becomes an effective experimental measure of the gap $\Delta(p)$ in the Fig.1a. In the following, substituting the ansatz $\psi = \psi_1 + \psi_2$ where $< \psi_1(\vec{x}) >= ae^{i\theta_1}$ and $< \psi_2(\vec{x}) >= e^{i\theta_2} \sum_{m=1}^{P} \Delta_m e^{i\vec{Q}_m \cdot \vec{x}}$ where $Q_m = G$ are the P shortest reciprocal lattice vectors of the lattice into Eqn.3, we study the effects of n lattice on ψ. From Eqn.3, we can see $n(x)$ acts as a periodic potential on ψ. In order to get the lowest energy ground state, we must consider the following 4 conditions: (1) because any complex ψ (up to a global phase) will lead to local supercurrents which is costy, so we can take ψ to be real, so $\vec{Q}_m$ have to be paired as anti-nodal points. P has to be even (2) as shown from the Feymann relation [11], $\vec{Q}_m, m = 1, \cdots, P$ are simply P shortest reciprocal lattice vectors, then translational symmetry of the lattice dictates that $\epsilon(\vec{K} = 0) = \epsilon(\vec{K} = \vec{Q}_m)$, ψ_1 and ψ_2 have to condense at the same time (3) The point group symmetry of the lattice dictates $\Delta_m = \Delta$ and is real (4) for the vacancies case, the interaction is attractive $g_v < 0$, from Eqn.3, we can see that the Superfluid Density wave (SDW) $\rho = |\psi|^2$ simple sits on top of the n lattice as much as possible. This is expected, because vacancies are hopping near the lattice sites, so the Bose condensation of vacancies also happen near the lattice sites. From Eqn.3, the attractive interaction also favors $\psi(x = \vec{R}/2) \sim 0$ where $\vec{R}/2$ stands for any interstitial sites which are in the middle of lattice points. It turns out that the the 4 conditions can fix the relative phase and magnitude of ψ_1 and ψ_2 to be $\theta_2 = \theta_1, \Delta = a/P$, namely:

$$\psi_{ss-v} = \psi_0 (1 + \frac{2}{P} \sum_{m=1}^{P/2} \cos \vec{Q}_m \cdot \vec{x}) \tag{7}$$

where $\psi_0 = ae^{i\theta}$ depends on the temperature and pressure. Note that in contrast to a uniform superfluid, the magnitude of ψ is changing in space. This field satisfies the Bloch theorem with the crystal momentum $\vec{k} = 0$ and the Fourier components are $\psi(\vec{K} = 0) = a, \psi(\vec{K} = \vec{Q}_m) = a/P$. They have the same sign and decay in magnitude. In principle, higher Fourier components may also exist, but they decay very rapidly, so can be neglected without affecting the physics qualitatively. $P = 6, 8, 12$ corresponds to sc, fcc, bcc lattices respectively [12].

5. Interstitial induced supersolid (SS-i): The discussion is quite similar to the vacancy case except that (1) if $g_i < g_{ic}$, the C-NS is an interstitial-like C-NS (named as NS-i) where the interstitial excitation energy ϵ_i is lower than that of the vacancy ϵ_v (2) if $g_i > g_{ic}$, the resulting solid is an in-commensurate solid with interstitials even at $T = 0$ whose condensation leads to $< \psi_i > \neq 0$, the SS-i state exists in the Fig.2. The arguments to determine the lattice structure of the SS-i goes the same as those in the vacancies case except the condition (4), for the SS-v, the interaction is attractive $g_v < 0$, so the SDW-v simply sits on top of the n lattice. However, for the interstitials case, the interaction is repulsive $g_i > 0$ which favors $\psi(x = 0) \sim 0$, so the Superfluid Density wave $\rho = |\psi|^2$ can avoid the n lattice as much as possible. It turns out that the the 4 conditions can fix the relative phase and magnitude of ψ_1 and ψ_2 to be $\theta_2 = \theta_1 + \pi, \Delta = a/P$, namely:

$$\psi_{ss-i} = \psi_0 (1 - \frac{2}{P} \sum_{m=1}^{P/2} \cos \vec{Q}_m \cdot \vec{x}) \tag{8}$$

where $\psi_0 = ae^{i\theta}$ depends on the temperature and pressure. Note that the crucial sign difference from the vacancies case which make the important difference in the X-ray scattering between SS-v and SS-i discussed in [11].

6. Low energy excitations in the SS: So far, we only look at the mean field solutions corresponding to vacancies and interstitials. Here we discuss excitations

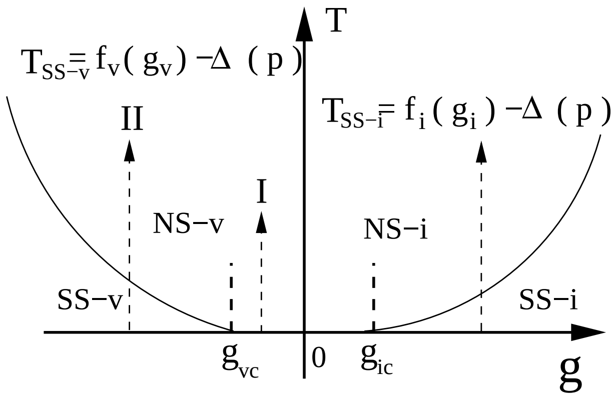

FIG. 2: The phase diagram of T versus g at a given pressure $p_{c1} < p < p_{c2}$. The finite temperature transitions denoted by the dashed line II at a given $|g_v| > |g_{vc}|$ (or $g_i > g_{ic}$) is in the 3D XY universality class. However, if $|g_v| < |g_{vc}|$, then the ground state at $T = 0$ is a vacancy-like C-NS(named as NS-v). Similar thing can be defined for an interstitial-like C-NS (named as NS-i) when $g_i < g_{ic}$.

above the mean field solutions. It turns out that the excitations in both cases are the same, so we discuss both cases at same time. In the SS-v and SS-i, the BEC wavefunctions can be written as

$$\psi_{ss} = \psi_0(1 \pm \frac{2}{P}\sum_{m=1}^{P/2} \cos \vec{Q}_m \cdot \vec{x}), \qquad \psi_0 = |\psi_0| e^{i\theta} \quad (9)$$

where $\pm$ sign corresponds to SS-v and SS-i respectively. Obviously, there are topological defects in the phase winding of θ which are vortices. At $T \ll T_{SS}$, the vortices can only appear in tightly bound pairs. However, as $T \rightarrow T_{SS}^-$, the vortices start to become liberated, this process renders $< \psi_0 >= 0$ above $T > T_{SS}$. In addition to the superfluid θ mode in the SS states, there are also lattice phonon modes $\vec{u}$ in both n sector and ψ sector. However, it is easy to see that the coupling Eqn.3 is invariant under $\vec{x} \rightarrow \vec{x} + \vec{u}, n(\vec{G}) \rightarrow n(\vec{G}) e^{i\vec{G}\cdot\vec{u}}, \psi(\vec{G}) \rightarrow \psi(\vec{G}) e^{i\vec{G}\cdot\vec{u}}$, so the lattice phonon modes in ψ are locked to those in the conventional n lattice. This is expected because there is only one kind of translational symmetry breaking, therefore only one kind of lattice phonons. *If ignoring the coupling between the superfluid θ mode and the phonon $\vec{u}$ mode* , then the action to describe the NS to SS transition in a static lattice is:

$$f_{\psi_0} = K_{NS} |\nabla \psi_0|^2 + t_{NS} |\psi_0|^2 + u_{NS} |\psi_0|^4 + \cdots \quad (10)$$

where $t_{NS} = T - T_{SS}$ and $T_{SS-v}(p) = f_v(g_v) - \Delta(p) = \Delta(p_{c2}) - \Delta(p)$ or $T_{SS-i}(p) = f_i(g_i) - \Delta(p) = \Delta(p_{c2}) - \Delta(p)$ (Fig.2). Obviously, it is still a 3d XY transition with a much narrower critical regime than the NL to SF transition [7]. It was shown in [6] that the coupling to the phonon mode $\vec{u}$ will not change the universality class at finite temperature. As the pressure increased to p_{c2}, T_{SS-v} or T_{SS-i} are suppressed to zero (Fig.1b), the system becomes a NS-v or NS-i where $< \psi_0 >= 0$. The zero temperature transition from the SS-v (SS-i) to the NS-v (NS-i) driven by the pressure near $p \sim p_{c2}$ will be studied in [11]. A low energy effective action involving the superfluid phonon θ, the lattice phonons $\vec{u}$ and their couplings will also be studied in [11].

7. Conclusions: By using the GL theory developed in [7], we analyze the microscopic mechanism and the conditions for the existence of a supersolid. There are two crucial parameters in the GL theory: v and g in Eqn.3. The v is an increasing function of the pressure p and determines the gap of the NS-PH, while the g is essentially a periodically changing chemical potential for the local superfluid mode ψ. In contrast to v, g is an intrinsic parameter of solid Helium 4 which depends on the mass of a ^4He atom and the potential between the ^4He atoms, but not sensitive to the pressure p. Taking the NS-PH state as the reference state, we show that a supersolid can be realized by adding small number of vacancies to the NS-PH. In fact, the temperature $T_{SS-v}(p) = \Delta(p_{c2}) - \Delta(p)$ determined in the experiments becomes an effective measure of the gap $\Delta(p)$ in Fig.1a). In [11], we will explore many important physical consequences due to this single coupling. Recently, in a Path integral quantum Monte-Carlo calculation, the authors in [5] concluded that the vacancies will simply move to the boundary and phase separate. This corresponds to the g lies in the NS-v regime in Fig.1 and Fig.2. However, our results do point out that there is a large parameter regime in Fig.1 and Fig.2 where the vacancy induced supersolid could be the favorable ground state. If the intrinsic parameter g does fall into this regime, then there is a NS-v to the SS-v transition described by Eqn.10. Indeed, very recent specific heat measurements found a excessive specific heat peak around the putative supersolid onset critical temperature $\sim 100\ mK$ [3]. More independent experiments are needed to see if g indeed falls into the regime, so the SS-v exists in ^4He.

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